Harmonic generation in ionizing systems by the complex scaled adiabatic-switch method

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An adiabatic-switch method for calculating harmonic generation under intense laser radiation is presented. The adiabatic switch is first applied to nonbound (i.e., ionizing or dissociative) time-periodic Hamiltonians when the energy spectrum is degenerate. Our derivation is based on the use of Floquet theory combined with complex scaling and of a generalized definition of the inner product for non-Hermitian operators. A model potential representing a Xe atom in the presence of a laser field is studied as an example to illustrate the numerical advantages of the present method and its stability.

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INTRODUCTION

High-harmonic generation (HG) (up to $\simeq 51$) has recently been detected in a system composed of rare-gas atoms in the presence of high-intensity laser fields [1]. Simulation methods based on a time-dependent Hartree-Fock approach have been successfully applied to the calculation of the harmonic-generation spectrum [2]. These simulations have clarified the distinction between the response of a single irradiated gas atom and the collective response of many atoms. Indeed, Potvliege and Shakeshaft [3] and DeVries [4] have calculated the harmonic generation spectrum for a single hydrogen atom in the presence of pulse laser. Moreover, the qualitative agreement between HG spectra which were obtained in simulations [5] and experimentally observed HG spectra had demonstrated that the HG can be studied by studying the dynamics of one-dimensional model Hamiltonians.

Recently [7] a simple time-independent expression for the probability to obtain high harmonics were derived by the use of Floquet theory combined with the complex-coordinate method and of the generalized definition of the inner product for non-Hermitian operators [6(a)]. The calculated HG spectra using the time-independent expression [given also in Eq. (1.10) in Sec. I was in complete agreement with the results obtained from motion of wave-packet calculations [7]. These calculations utilized the time periodicity of the laser source and the complex-coordinate method [6] in order to analyze the system in terms of quasienergy (Floquet [8]) resonance states. Calculations were carried out by the time-dependent complex-coordinate Floquet method [9] (TDCCFM) (with the use of a Fourier grid representation) which requires the solution of the evolution equation to obtain the evolution matrix for one optical cycle, and the subsequent diagonalization of that matrix to obtain the eigenvectors associated with the quasienergy resonance states.

The first step in the time-dependent complex-coordinate Floquet method involves the propagation of a matrix such that the computational effort scales as $N^2 \ln N$ [if fast-Fourier-transform (FFT) representation is used] where N is the dimension of the matrix. The diagonalization which is required in the second step scales as N^3. For the one-dimensional model Hamiltonian which was used in Ref. [7], where a faithful representation in Hilbert space required not more than $N=512$ Fourier grid points, it turned out that the propagation was the rate-limiting step. For more realistic model Hamiltonians having more than one degree of freedom, the computational effort will eventually be determined by diagonalization.

Assuming that the dynamics of the system is dominated by the quasienergy resonance state with the largest lifetime (as was done in our calculations), it is a waste of computational effort to propagate *all* of the basis vectors. In Sec. I we present an alternative approach for this problem. This approach is based on the complex-coordinate method which enables us to isolate a resonance state from the other states in the continuum and to remove the degeneracy from the problem. Only because of these facts can one turn on the external field adiabatically during time propagation of a field-free initial state. One can choose the initial state such that the resonance obtained is the one with the largest lifetime. The present approach involves the propagation of a single *vector* (in comparison with the time-dependent complex-coordinate Floquet method which requires the propagation of a *matrix*) such that the computational effort scales as $N \ln N$.

In Sec. I the complex scaled adiabatic-switch method (CSASM) is presented. In Sec. II a model Hamiltonian for the ionization of a Xe electron will be introduced. Illustrative numerical results will be presented and compared to results obtained by solving the complex scaled time-dependent evolution equation [7] in Sec. III. The final section will conclude.

I. HARMONIC GENERATION BY THE COMPLEX SCALED ADIABATIC-SWITCH METHOD

The creation of harmonic generation (as well as other nonlinear effects which are observed when placing atoms

in strong laser fields) can be explained in terms of the interaction of a *single* electron with its nucleus and the external driving force [2]. Furthermore, we recently demonstrated that HG spectra are accurately obtained when the pulse laser is approximated as a *periodic* source [7]. With this approximation, the Hamiltonian (in radiation gauge, and for $\hbar=m=1$) can be written as

$$\hat{H}_{(x,t)}=\frac{1}{2}\left[\hat{p}-\frac{E_0}{\omega}\sin(\omega t)\right]^2+V(x)\ , \tag{1.1}$$

where E_0 is the field strength parameter. The advantage of such a Hamiltonian is its time periodicity which allows solution, $\psi(x,t)$, of the time-dependent Schrödinger equation

$$i\frac{\partial\psi(x,t)}{\partial t}=\hat{H}(x,t)\psi(x,t) \tag{1.2}$$

in terms of Floquet states [8]

$$\Phi_\alpha(x,t)=\Phi_\alpha(x,t+nT)\ ,\quad T=\frac{2\pi}{w}\ ,$$

and quasienergies ϵ_α

$$\psi(x,t)=\sum_\alpha C_\alpha\Phi_\alpha(x,t)\exp^{-i\epsilon_\alpha t}\ . \tag{1.3}$$

Complex scaling of the coordinate $x\rightarrow xe^{i\theta}$, divides the quasienergy spectrum, ϵ_α, into two groups [9] as follows.

(I) Continuum states: $|\epsilon_\alpha-\omega n|e^{2i\theta}$; $n=-\infty,\ldots,\infty$ when ωn are the threshold energies. The rotating "white" continua do not contain any information about the resonance phenomena.

(II) Resonance states: $\epsilon_\alpha=\epsilon_\alpha(\text{pos})-i\Gamma_\alpha/2$ having positions, $\epsilon_\alpha(\text{pos})$, and finite widths $\Gamma_\alpha=\hbar/\tau_\alpha$ in the complex energy plane. The width is inversely proportional to the lifetime, τ_α. The discrete resonance states are θ independent provided that θ is sufficiently large.

For sufficiently long evolution times (and for large enough rotational angle θ) only the quasienergy resonance state that has the *largest lifetime*—the narrowest width—survives. Under such assumptions the sum in Eq. (1.3) can be reduced to

$$\psi(xe^{i\theta},t)\simeq\Phi^{\text{res}}(xe^{i\theta},t)\exp\left[-i\left[\epsilon_{\text{res}}(\text{pos})-i\frac{\Gamma_{\text{res}}}{2}\right]t\right]\ . \tag{1.4}$$

Prior to the application of the external field, the atom is assumed to be in its ground state. Switching the field on transforms this ground state into a metastable one. This ground state will appear as the resonance state with the largest lifetime (see, for example, the results presented in Table 8 in Ref. [10]). It is therefore argued that in many cases the dynamics of the system can be adequately described by investigating the time evolution of this single state. (Chu and Cooper [11] and later on Szöke [12] had theoretically discussed this assumption and its limitations.) We therefore propose a procedure by which this state is propagated in time as the external field is adiabatically switched on. The idea of carrying out an adiabatic switch, where E_0 becomes time dependent, is an old one, and a good set of references for it appears in the paper by Reinhardt and Dana [13]. Adiabatic switching is also discussed in the context of periodic systems by Potvliege and Shakeshaft [8(f)] and by Breuer, Dietz, and Holthaus [14]. A formal proof of the adiabatic theorem for the case of a periodic Hamiltonian has been presented by Young and Deal [15]. Hence we solve the time-dependent Schrödinger equation (1.2) with the Hamiltonian given by Eq. (1.1) where the field intensity parameter varies from 0 to its maximal value, E_0, as time passes and at $t=0$ the system is described by $\psi(xe^{i\theta},t=0)$ which is the ground state of the field-free Hamiltonian.

Having obtained $\psi(xe^{i\theta},t)$, it is possible to calculate the harmonic-generation spectrum (as well as the above-threshold ionization spectrum and other properties) that is defined as

$$\sigma(\Omega)\propto\left|\frac{1}{T}\int_0^T e^{-i\Omega t}D(t)dt\right|^2\ , \tag{1.5}$$

where

$$D(t)=\langle\langle\,\psi(xe^{i\theta},t)|\hat{p}|\psi(xe^{i\theta},t)\,\rangle\rangle \tag{1.6}$$

is calculated for one optical cycle $T=2\pi/\omega$. The inner product $\langle\langle\ \rangle\rangle$ stands for the *complex* product described in Ref. [6(a)] (i.e., no complex conjugation of terms which are complex as a result of scaling).

For calculating the complex inner product, $D(t)$, in Eq. (1.6), it is recommended to represent $\Phi^{\text{res}}(xe^{i\theta},t)$ in Eq. (1.4) as a Fourier series (using the time periodicity of $\Phi(xe^{i\theta},t)$ explicitly),

$$\begin{aligned}|\psi_{\text{res}}\rangle\rangle&=e^{-i\lambda_{\text{res}}t}|\Phi^{\text{res}}(t)\rangle\rangle\ ,\\ \langle\langle\,\psi_{\text{res}}|&=e^{+i\lambda_{\text{res}}t}\langle\langle\,\Phi^{\text{res}}(t)|\ ,\end{aligned} \tag{1.7}$$

where

$$\begin{aligned}|\Phi^{\text{res}}(t)\rangle\rangle&=\sum_{k=-\infty}^{\infty}|\phi_k^{\text{res}}(\theta)\rangle\rangle e^{+iwkt}\ ,\\ \langle\langle\,\Phi^{\text{res}}(t)|&=\sum_{k=-\infty}^{\infty}\langle\langle\,\phi_k^{\text{res}}(\theta)|e^{-iwkt}\ .\end{aligned} \tag{1.8}$$

Since in our studied case the Floquet Hamiltonian matrix, $\mathcal{H}$, is a symmetric one, and since $|\phi_k^{\text{res}}\rangle\rangle$ and $\langle\langle\,\phi_k^{\text{res}}|$ are, respectively, the right and left eigenvectors of $\mathcal{H}$ then

$$\langle\langle\,\phi_k^{\text{res}}(\theta)|=|\phi_k^{res}(\theta)\rangle\rangle=\phi_k^{\text{res}}(xe^{i\theta})\ . \tag{1.9}$$

As θ trends to zero $\{\phi_k^{\text{res}}(x)\}$ get real values only. Therefore $\phi_k^{\text{res}}=(\phi_k^{\text{res}})^*$ and Eq. (1.6) reduces to the usual definition of $D(t)=\langle\,\psi(t)|\hat{p}|\psi(t)\,\rangle$.

Substitution of Eqs. (1.7)–(1.9) to Eqs. (1.5), (1.6), and (1.4) leads to the final result for the probability of the system to emit radiation in frequency $\Omega=n\omega$ (see also Ref. [7])

$$\sigma(\Omega=n\omega)\propto\left|\sum_{k=-\infty}^{\infty}\langle\langle\,\phi_{k+n}^{\text{res}}(\theta)|\hat{p}|\phi_k^{\text{res}}(\theta)\rangle\rangle\right|^2\ . \tag{1.10}$$

This time-independent expression was obtained due to the use of the generalized definition of the inner product for non-Hermitian operators [6(a)]. To sum up we outline the proposed procedure as follows.

(i) The time-dependent Schrödinger equation (1.2) for the (given) Hamiltonian (1.1) is solved when the lowest ground state of the field-free Hamiltonian is taken as an initial state. During the time propagation, the external field strength parameter is initialized at zero and is adiabatically increased to its maximal value, E_0.

(ii) For specific values of the field strength parameter, E_0,

$$\psi_{\rm res}(xe^{i\theta},t)=\Phi^{\rm res}(xe^{i\theta},t)\exp\left[-i\left[\epsilon_{\rm res}({\rm pos})-i\frac{\Gamma_{\rm res}}{2}\right]t\right] \tag{1.11}$$

is propagated for one more optical cycle to give

$$\psi_{\rm res}(xe^{i\theta},t+T)=\Phi^{\rm res}(xe^{i\theta},t+T)\exp\left[-i\left[\epsilon_{\rm res}({\rm pos})-i\frac{\Gamma_{\rm res}}{2}\right](t+T)\right] . \tag{1.12}$$

Dividing $\psi_{\rm res}(xe^{i\theta},t+T)$ by $\psi_{\rm res}(xe^{i\theta},t)$ and taking the periodicity of Φ into consideration one obtains the Floquet exponent $\lambda_{\rm res}=\exp[-i(\epsilon_{res}({\rm pos})-i\Gamma_{\rm res}/2)T]$.

(iii) Propagating $\psi_{\rm res}(xe^{i\theta},t)$ for one more optical cycle for the chosen fixed field strength parameter, E_0, and using the known Floquet exponent, $\lambda_{\rm res}$, the Fourier components $\phi_k^{\rm res}(xe^{i\theta})$ for each time step are calculated. That is,

$$\phi_k^{\rm res}(xe^{i\theta})=\frac{1}{T}\int_0^T e^{-i\omega kt}\psi_{\rm res}(xe^{i\theta},t)\Big/(\lambda_{\rm res})^{t/T}dt . \tag{1.13}$$

(iv) $\sigma(\Omega=n\omega)$ is calculated by using the Fourier components $\phi_k^{\rm res}(xe^{i\theta})$ via Eq. (1.8).

II. AN ILLUSTRATIVE NUMERICAL EXAMPLE

An inverse Gaussian potential was suggested by Bardsley, Szöke, and Comela [16] for describing the interaction of a Xe electron with an external laser field,

$$V(x)=-V_0\exp[-(x/x_0)^2] . \tag{2.1}$$

For $V_0=0.63$ a.u. and $x_0=2.65$ a.u., the field-free potential supports two bound states which mimic the two lowest electronic states of Xe, $E_0=-0.4451$ a.u. and $E_1=-0.1400$ a.u., and a third weakly bound state with energy of $E_2=-0.000\,14$ a.u. The external field frequency in Eq. (1.1) was set very close to a three-photon resonance $\omega=0.0925$ a.u. and the maximum field intensity $I_0=(c/8\pi)E_0^2$ was 10^{14} W/cm^2 in most of the calculations.

As mentioned above, an adiabatic switch of the field intensity was used. Theoretically a smooth function should have been used [15]. Practically there was no numerical difference between a linear switch [i.e., $E_0(t)=bt$, where b is a small constant], and a cosine switch. This might be due to the $\sin(\omega t)$ prefactor in the Hamiltonian, Eq. (1.1), that had smoothed the sharp edges of $E_0(t)$ at the initial and final times of the turning-on process because the turning-on time was set to integer multiples of the external field period, T. Typically, a switch of $I_0=10^{13}$ W/cm^2 required 10–100 optical cycles. The coordinate was complex scaled by a nonphysical parameter θ such that $x\rightarrow xe^{i\theta}$. A typical value of $\theta=0.5$ rad was used for most of the calculations. The stability of such calculations with respect to changes in the nonphysical parameter θ is demonstrated elsewhere [7,9(b)].

Methods for solving the Schrödinger equation (1.2) using the time-dependant complex-coordinate Floquet method are described in detail in Ref. [9(a)]. In this work the Hamiltonian and wave packet were represented in a Fourier grid (typically 256 grid points with a grid step of 0.8 a.u. were used). Solution of the Schrödinger equation (1.2) was carried out in the integral form, where the evolution matrix was evaluated at small time steps dt (a typical one being $T/4096$) for which the Hamiltonian was assumed to be almost constant. $\hat{T}\exp[-i\int H(t)dt]$, where $\hat{T}$ is the time-ordering operator, was approximated by a second-order Magnus series [17]. Matrices exponentiation was carried out by Taylor expansion of fifth to tenth order. Operation of the Hamiltonian on the wave packet was carried out for the kinetic part and for the potential part separately. Forwards and backwards FFT were used for transforming the wave packet from momentum space to configuration space and back [18]. Complex scaling induces decay of the amplitude of the wave packet during the time propagation. In order to avoid dealing with very small numbers, the wave packet was renormalized to an arbitrary value (1.0) after each optical cycle.

III. RESULTS

In previous studies [7,9], we demonstrated the advantages in the representation of the Floquet characteristic resonances, $\lambda_{\rm res}=\exp[-i(\epsilon_{\rm res}({\rm pos})-i\Gamma_{\rm res}/2)T]$ on the complex λ plane instead of examining the actual complex resonance eigenvalues $[\epsilon_{\rm res}({\rm pos})-i\Gamma_{\rm res}/2]$. In such a representation, the "distance" of a resonance from the origin (0,0) is proportional to its lifetime such that bound states with infinite lifetimes are situated on the unit circle curve, while resonances with short lifetimes are situated closer to the origin (0,0). The angle in this complex plane describes the energy position (modulus ω). The dependence of $\lambda_{\rm res}$ of the resonance correlated with the ground

state on the intensity of the field, I_0, is shown in Fig. 1. Figure 1(a) shows the results obtained by the complex scaled adiabatic-switch method, (2048 time steps per optical cycles, and 100 optical cycles for switching on I_0 by 10^{13} W/cm^2 in a cosine way). Figure 1(b) shows the results obtained (for the same model) by diagonalizing the whole complex scaled time evolution matrix calculated by the time-dependent complex-coordinate Floquet method [7] for one optical cycle.

Comparing the two figures, one can clearly see the agreement for field intensities in the range from $I_0=0.0$ to 22×10^{13} W/cm^2. The slight "swerve" around $I_0=12.5\times10^{13}$ W/cm^2 in Fig. 1(b) is a direct result of an avoided-crossing event of the ground resonance state (shown) and another resonance state [see Fig. 2(b) of Ref. (7)]. Following an avoided-crossing event in an adiabatic way [Fig. 1(a)] requires a gradual switching on the field. Indeed, for example, when the field intensity was switched on 10 times faster (i.e., 10 optical cycles per switch of 10^{13} W/cm^2) we were not able to obtain the correct results around the first avoided-crossing event at $I_0=12.5\times10^{13}$ W/cm^2.

For intensities higher than $I_0=22\times10^{13}$ W/cm^2, there are some discrepancies between Figs. 1(a) and 1(b). A very drastic avoided-crossing event appears around $I_0=22.5\times10^{13}$ w/cm^2 [see Fig. 2(b) of Ref. [7]]. In this case the ground-state resonance undergoes two avoided-crossing events with other resonance states, one followed by another. These frequent avoided-crossing events cause the ground-state resonance to undergo a 180° turn in the complex λ plane [Fig. 1(b)]. This sequence of events is too drastic to be followed even by the gradual adiabatic switch used here. Presumably, this avoided-crossing event can also be traced by a more gradual switching procedure.

The harmonic-generation spectrum obtained for $I_0=10^{14}$ W/cm^2 is presented in Fig. 2. These results are in perfect agreement with results obtained by diagonalization of the complex scaled time evolution matrix calculated by the time-dependent complex-coordinate Floquet method [7], for one optical cycle. Since the potential is symmetric (parity is a good quantum number), and since the dipole approximation was used, even harmonics are suppressed [19]. This phenomenon is clearly seen in the figure. Looking at the odd harmonics, one can see an exponential decay (note the logarithmic scale) from the first

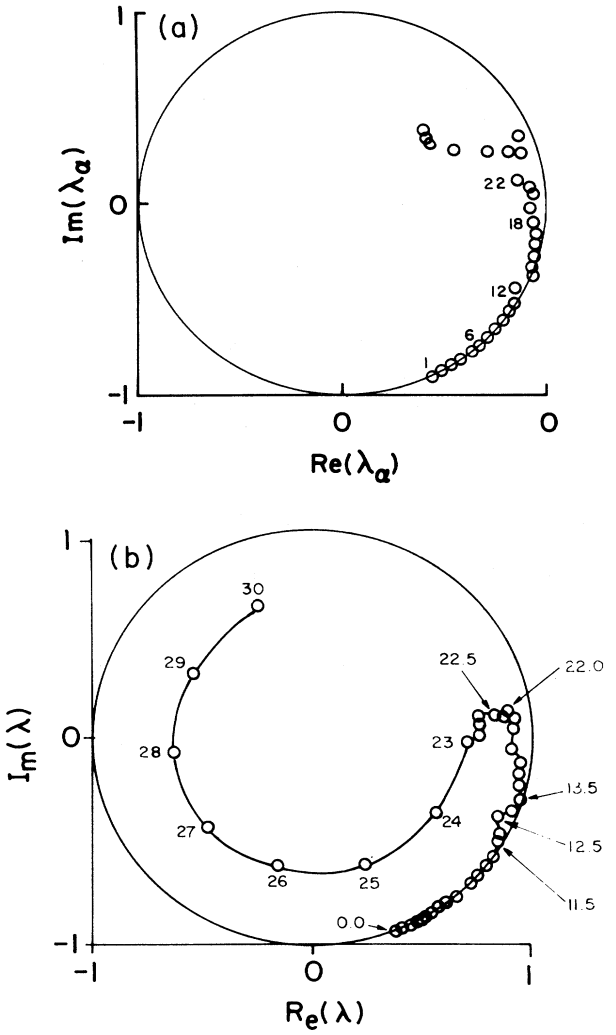

FIG. 1. Dependence of $\lambda_{res}=\exp[-i(\epsilon_{res}(\text{pos})-i\Gamma_{res}/2)T]$ on the maximum field intensity, I_0, for the model Hamiltonian in Eqs. (1.1) and (2.1) ($\omega=0.0925$ a.u., $V_0=0.63$ a.u., $X_0=2.65$ a.u.). The field intensities $I_0=(c/8\pi)\epsilon_0^2$ are given in units of 10^{13} W/cm^2. The results presented in (a) were calculated by the CSASM with a cosine switch (2048 time steps per optical cycle, 100 optical cycles per switching I_0 by 10^{13} w/cm^2), and those presented in (b) were calculated by the TDCCFM [7].

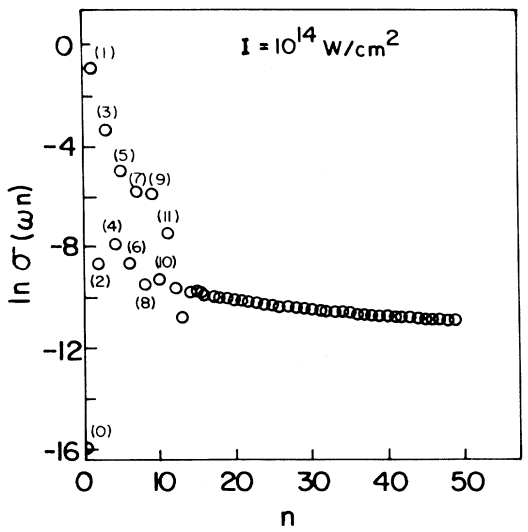

FIG. 2. Harmonic-generation spectrum: $\ln\sigma(n\omega)$ vs $n=\Omega/\omega$ [Eq. (1.8)] calculated by the CSASM for the model Hamiltonian given in Eqs. (1.1) and (2.1) for $I_0=10^{14}$ W/cm^2. A linear switch was used (4096 time steps per optical cycle, 10 optical cycles per switch of 10^{13} W/cm^2), 256 Fourier grid points with a grid step of 0.78 a.u.

harmonics to the fifth one, followed by a (short) plateau between the fifth harmonics and the ninth harmonics. This plateau ends up in a final exponential drop. The same qualitative behavior (i.e., an exponential decay to a plateau that ends up in a final drop) was also detected experimentally [1].

CONCLUDING REMARKS

A quantum-mechanical method (complex scaled adiabatic-switch method, CSASM) for calculating harmonic generation was presented. The backbone of the method is the adiabatic theorem. This theorem is phrased for a Hamiltonian which depends on time only through the adiabatic parameter (i.e., nonexplicitly), and which has a nondegenerate energy spectrum. For such Hamiltonian the Gell-Mann and Low theorem [20,21] states that by adiabatically (slowly, $b \rightarrow 0$) turning a perturbation on, the ground state of some $\hat{H}_0$ Hamiltonian at time $t = -\infty$ evolves in such a way that at time $t = 0$ it becomes an eigenstate (not necessarily the lowest one) of another Hamiltonian $\hat{H} = \hat{H}_0 + \exp(-b|t|)\hat{H}_1$.

In our case the Hamiltonian, Eq. (1.1), has an explicit dependence on time, and all resonance states are embedded in a *degenerate* continuum. *A priori* it seems inadequate to apply a computational scheme which is based on an adiabatic switch, because of these two difficulties.

For time-periodic Hamiltonians, such as the Hamiltonian given in Eq. (1.1), and for a gradual enough switch of the external field, the ground state of the field-free Hamiltonian (at time $t = 0$) is forced to stay on a single *quasienergy* shell. Therefore, as proved by Young and Deal [15], one is allowed to use the adiabatic switch for this form of time dependence, which lies beyond the conventional range of applicability of the Gell-Mann and Low theorem, such that the first difficulty in applying the adiabatic switch to our system is overcome. The use of *complex scaling* separates between the resonance states and the continuum states [22] such that the degeneracy is removed, and the second difficulty is overcome.

The numerical results obtained by the complex scaled adiabatic-switch method were in agreement with the results obtained by an accurate numerical method [7] which requires a time propagation of the time evolution *matrix* for one period. Here only *one vector* is propagated so the computational effort usually reduces drastically. The adiabatic switch is, therefore, a promising solution for problems with several degrees of freedom. The procedure has a disadvantage in describing an avoided-crossing event in the complex λ plane, since one observes only one branch which is connected with a single resonance state. The full avoided-crossing picture will be obtained by repeating the adiabatic-switch calculation when different eigenstates of the field-free Hamiltonian are taken as initial states.

Here we used the complex scaled adiabatic-switch method to study the harmonic-generation spectrum. Other interesting multiphoton phenomena observed for atoms or molecules interacting with intense laser fields (such as above-threshold ionization or dissociation) can be treated by this method as well.

It is worthwhile to consider the issue of relating the Floquet analysis which is basically a cw approach to the short pulse experiments which are required to obtain the high field intensity. Two extreme situations can be encountered depending on the rate of the turnon of the pulse. In the sudden limit the pulse will create a superposition of resonances and continuum states. Then as time passes the continuum states will scatter off, followed by the short living resonances. Due to the high kinetic energy acquired by the electrons they move fast out of the interaction region. This means that after a short induction period the longest living resonance will dominate the dynamics. The other extreme is a pulse with the adiabatic turnon. In this case the system will "stay" on the resonance state which is correlated with the ground state of the field-free Hamiltonian.

In the sudden-limit approximation the complex scaled adiabatic-switch method suggests a representation of the *real* system up to field intensities where an avoided-crossing event causes a drastic decrease in the lifetime of the resonance state correlated with the ground state of the field-free system ($I_0 = 22.5 \times 10^{13}$ W/cm^2 in our case). In the other extreme case the complex scaled adiabatic-switch method suggests a *precise* way to follow the experimental reality.

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